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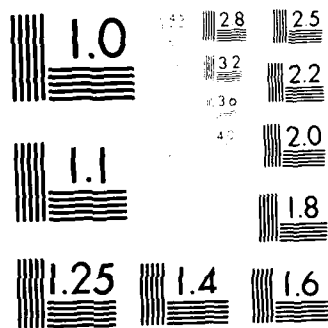
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ANNEALING EFFECTS IN FERROMAGNETIC AMORPHOUS ALLOYS

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## 1. INTRODUCTION

Ferromagnetic amorphous alloys, or metallic glasses, have been under development as engineering materials for about ten years [1]. Cobalt base zero-magnetostrictive alloys are already being used for high permeability application such as recording head, and iron-nickel base alloys are beginning to be used in switching regulator. Large-scale application in power transformers is now seriously considered.

In all these applications, heat treatment or annealing is an important, indispensable step. Metallic glasses are usually produced by rapid quenching process. During this process often some internal stresses are frozen in, and also stress-induced magnetic anisotropy can be produced. Since these by-products are usually undesirable for application, they are removed by appropriate annealing treatment. Furthermore, as-quenched alloys retain "high temperature structure", which are not stable at low temperatures, thus annealing is necessary to stabilize the structure. However, the effects of annealing are very complex and multi-faceted, and scientific understanding of this important process has been quite unsatisfactory. The purpose of this project is to examine various annealing effects carefully, and to arrive at a more complete microscopic and quantitative description of the process.

During the four years of funding, we studied the annealing effects on permeability, field-induced magnetic anisotropy, magnetic after-effects, domain structure, Curie temperature, anelasticity and elasticity, and creep-induced magnetic anisotropy. We have used not only magnetic measurement techniques, but also energy-dispersive x-ray diffraction, internal friction and mechanical creep measurements, and computer modeling and simulation to analyze these effects. We found that all the reversible relaxation processes are closely connected, sharing kinetics and microscopic processes, and obtained a quantitative description of the annealing kinetics, including the effect of pre-annealing conditions on the relaxation kinetics. Also the atomic level processes are now much better understood through the study of anelasticity. These results have been, or are going to be, published in more than 30 scientific publications which include several invited review papers. Thus the goals of the project have been successfully achieved.

## 2. RESEARCH RESULTS FOR THIS REPORTING PERIOD.

### 2.1 Microscopic Process of Anelastic Relaxation.

We have succeeded in obtaining the first direct evidence of the structural anisotropy associated with anelastic deformation. Using the energy-dispersive x-ray diffraction (EDXD) technique [2] which offers very high accuracy of structural determination for amorphous solids, the anisotropic component of the atomic pair distribution function was determined before and after the anelastic deformation by mechanical creep. It was found that the nearest neighbour bond reorientation produces the anelastic polarization.

This observation has several important implications with regard to the microscopic processes of anelastic deformation:

1. The relaxation centers, or the structural defects, are very much localized to within one or two atomic distances.
2. The density of the relaxation centers is of the order of the anelastic strain, i.e. the local strain times the volume of the defect, activation volume, is of the order of the atomic volume. This result agrees with the macroscopic observation of the activation volume.
3. The anelastic polarization is recovered by the subsequent annealing without load.
4. The phenomenological picture of independent relaxation centers is fully justified.

The result is also consistent with the result of computer simulation which identified several modes of local bond reorientation to produce anelastic strain. A paper describing the details of these processes is under preparation. As our early investigations have proven, the anelastic relaxation provides the mechanism of reversible relaxation phenomena, including various magnetic phenomena. Therefore these results are directly related to the main subject of this project.

## 2.2 Kinetics of the Reversible Structural Relaxation Phenomena.

Annealing effects on magnetic anisotropy and Curie temperature are largely reversible, but appear to contain also some irreversibility. In particular, the kinetics of the change become slower after annealing, and this has been explained by many researchers in terms of "annihilation of free volumes" which causes the increase in viscosity. We have shown that this common explanation is in fact incorrect, and suggested a more complete quantitative description of the phenomena.

Through the detailed study of anelasticity, we have determined the activation energy spectrum for relaxation centers [3]. The spectrum is also a function of the "fictive temperature", or the temperature at which the present configuration will be in thermal equilibrium. In the as-quenched sample, for example, the fictive temperature is usually quite high, often higher than the glass transition temperature by 200-400 K. By annealing at a lower temperature for a long time, this fictive temperature  $T_f$  can be brought to that annealing temperature. This dependence on  $T_f$  describes the effect of the pre-annealing condition on the kinetics of relaxation.

We have shown that the kinetics of the field-induced magnetic anisotropy  $K_u$  and the change in the Curie temperature  $T_c$  can be very well explained, including the effect of the pre-annealing conditions, by the activation energy spectrum for anelasticity except that they saturate much more quickly; the kinetics of  $K_u$  and  $T_c$  require only the lowest energy portion of the spectrum which is about 5-10% of the total. This observation indicates that an anelastic relaxation event changes the local compositional short range order of about ten atoms, which is indeed consistent with the observation by computer simulation.

We have also determined the kinetics of the change in  $T_f$ , or the kinetics of the change in the kinetics. The activation energy for these kinetics,  $E_k$ , which is the activation energy to change the density of the activation energy spectrum at  $E_a$ , was found to be, somewhat surprisingly, proportional to  $E_a$ , with the average value of about  $1.2 \times E_a$ . For Fe(40)Ni(40)P(14)B(6) alloy, the activation energy spectrum has the following ranges;

- 1 - 2.2 eV: Magnetic relaxation, for  $K_u$  and  $T_c$
- 1 - 2.6 eV: Activation energy for fictive temperature
- 1 - 2.8 eV: Anelasticity
- above 2.8 eV: Isoconfigurational flow, viscosity

Thus the microscopic processes for magnetic relaxation and those for viscous flow cannot be the same, since they have different activation energies. If the viscous flow occur due to the free volume diffusion, the magnetic relaxation processes must occur for a different mechanism. We propose that it is the diffusionless shear transformations.

### 2.3 PUBLICATIONS

The results summarized above are described in more detail in the publications listed separately. In the list no. 19-29 were either published or submitted for publication during the report period.

### 3. SUMMARY OF THE PROJECT

The present project carried out over about four years has yielded the following conclusions regarding the annealing effects on ferromagnetic amorphous alloys.

1. Annealing the ferromagnetic amorphous alloys produces structural relaxation which affects most of the physical properties. The manner with which the annealing affects the property, however, is not the same for all the properties. In some cases the effect is irreversible below  $T_g$ , while in other cases it is clearly reversible. Magnetic properties such as the field-induced anisotropy  $K_u$ , creep-induced anisotropy  $K_s$ , Curie temperature  $T_c$ , and the permeability disaccommodation  $DA$ , are all reversible; they saturate to temperature dependent equilibrium values after some time during annealing, and if the annealing temperature is reversibly changed, they follow the change with some characteristic delay. We have shown that the four magnetic properties listed above and anelasticity measured by internal friction all share the basically same kinetics, and therefore the same microscopic mechanism.
2. An analytical expression for this reversible relaxation behavior has been phenomenologically determined. The expression also includes the effect of the pre-annealing condition, or the fictive temperature of the system.

3. By computer simulation and x-ray diffraction, the microscopic mechanism of the reversible relaxation phenomena has been determined to be the local shear deformations which re-orient some of the atomic bonds.

#### REFERENCES

1. T. Egami, Rep. Prog. Phys. 47, 1601 (1984).
2. T. Egami, in "Metallic Glasses I", ed. H. J. Guntherodt and H. Beck (Springer Verlag, Berlin, 1981), p. 25.
3. N. Morito and T. Egami, Acta Metall. 32, 603 (1984).

Publications from Previous Contract  
N00014-80-C-0896; NR 039-204

1. Structure and Magnetism of Amorphous Alloys  
T. Egami  
IEEE Trans. Magn. MAG-17 (1981) 2600.  
(Invited paper at INTERMAG, Grenoble, France)
2. Kinetics of Formation of Induced Magnetic Anisotropy in a Zero-Magnetostriction Amorphous Alloy  
Kai-Yuan Ho, P. J. Flanders, and C. D. Graham, Jr.  
J. Appl. Phys. 53 (1982) 2279.
3. Physical Origin of Losses in Conducting Ferromagnetic Materials  
C. D. Graham, Jr.  
J. Appl. Phys. 53 (1982) 8276.  
(Invited paper at Conf. on Magnetism and Magnetic Materials, Montreal)
4. Isotropic Behavior of the Kinetics of Reorientation of Induced Anisotropy in Amorphous and Crystalline Alloys  
Kai-Yuan Ho  
J. Appl. Phys. 53 (1982) 7828.
5. Kinetics of Reorientation of Induced Anisotropy in Amorphous and Crystalline Alloys  
Kai-Yuan Ho  
J. Appl. Phys. 53 (1982) 7831.
6. Kinetics of Changes in Initial Permeability Produced by Magnetic Annealing in a Zero-Magnetostrictive FeCoSiB Amorphous Alloy  
T. Jagielinski  
J. Appl. Phys. 53 (1982) 7855.
7. Elimination of Disaccommodation in a Zero-Magnetostrictive FeCoSiB Amorphous Alloy  
T. Jagielinski  
J. Appl. Phys. 53 (1982) 7852.
8. Structural Relaxation and Magnetism in Amorphous Alloys  
T. Egami  
J. Mag. Magn. Mat. 31-34 (1983) 71.  
(Invited paper, International Conf. Magnetism, Kyoto)
9. Single-Ion Anisotropy and Magnetostriction of Amorphous Alloys  
Y. Suzuki and T. Egami  
J. Mag. Magn. Mat. 31-34 (1983) 1549.
10. Internal Friction of a Glassy Metal  $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$   
During the Cross-Over Behavior of the Curie Temperature  
N. Morito and T. Egami  
IEEE Trans. Magn. MAG-19 (1983) 1898.



11. Correlation Between the Changes Due to Heat Treatment in the Curie Temperature and Internal Friction of a Glassy Metal  
 $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$   
N. Morito and T. Egami  
IEEE Trans. Magn. MAG-19 (1983) 1901.
12. Field Induced Anisotropy in Zero Magnetostriction Amorphous Alloys Measured with a Rotating Sample Magnetometer  
P. J. Flanders, T. Egami, and C. D. Graham, Jr.  
IEEE Trans. Magn. MAG-19 (1983) 1904.
13. The Relationship Between Changes in Field-Induced Anisotropy and Curie Temperature for  $\text{Fe}_{40}\text{Ni}_{40}\text{B}_{14}\text{P}_6$   
P. J. Flanders, N. Morito, and T. Egami  
IEEE Trans. Magn. MAG-19 (1983) 1907.
14. Annealing Kinetics for Curie Temperature Changes in the Amorphous Alloy  $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$   
P. J. Flanders, N. Morito, and T. Egami  
IEEE Trans. Magn. MAG-19 (1983) 1910.
15. Effects of Anisotropy on Domain Structure in Amorphous Alloys  
J. D. Livingston, W. G. Morris, and T. Jagielinski  
IEEE Trans. Magn. MAG-19 (1983) 1916.
16. Flash Annealing of Amorphous Alloys  
T. Jagielinski  
IEEE Trans. Magn. MAG-19 (1983) 1925.
17. Disaccommodation of Magnetic Permeability in Amorphous Iron-Nickel-Boron Alloys  
T. Jagielinski  
IEEE Trans. Magn. MAG-19 (1983) 1934.
18. Elastic Stress-Induced Coercive Field Changes in NiCo Films Used in a Rotating Disk  
P. J. Flanders  
IEEE Trans. Magn. MAG-19 (1983) 1680.
19. Internal Friction and Reversible Structural Relaxation in Metallic Glass  $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$   
N. Morito and T. Egami  
Acta Met. 32 (1984) 603.
20. Correlation of Shear Modulus and Internal Friction in the Reversible Structural Relaxation of a Glassy Metal  
N. Morito and T. Egami  
J. Non-Cryst. Solids 61/62 (1984) 973.

21. Reversibility of the Structural Relaxation in Amorphous Alloys  
T. Jagielinski and T. Egami  
J. Appl. Phys. 55 (1984) 1811.
22. Effects of Applied Currents on Domain Structures and Permeability  
in Amorphous Metal Ribbons  
J. D. Livingston, W. G. Morris, and T. Jagielinski  
J. Appl. Phys. 55 (1984) 1790.
23. The Effect of Flash Annealing on the Magnetic Properties of  
Amorphous Alloys (Abstract)  
T. Jagielinski  
J. Appl. Phys. 55 (1984) 1799.
24. Kinetics of the Reversible Relaxation Phenomena in Metallic  
Glasses  
T. Egami and T. Jagielinski  
to be published in Proc. Int. Conf. Rapidly-Quenched Metals 5  
(1984).
25. Distributed Fictive Temperatures and the Kinetics of Structural  
Relaxation  
T. Egami  
Invited review, to be published in Proc. Int. Conf. Rapidly-  
Quenched Metals 5 (1984).
26. Kinetics of the Curie Temperature Relaxation and Anelasticity  
in Amorphous Alloys  
T. Jagielinski and T. Egami  
J. Appl. Phys. 56 (1985) to be published.
27. Magnetic and Structural Effects of Anelastic Deformation  
of an Amorphous Alloy  
J. Haimovich, T. Jagielinski and T. Egami  
J. Appl. Phys. 56 (1985) to be published.
28. Kinetics of the Fictive Temperature during the Relaxation  
of Field-Induced Anisotropy in Amorphous Alloys  
T. Jagielinski and T. Egami  
Abstract submitted for INTERMAG 85.
29. Creep-Induced Magnetic Anisotropy in Amorphous Alloys:  
Kinetics and Equilibrium Values  
T. Jagielinski and T. Egami  
Abstract submitted for INTERMAG 85.
30. Direct Observation of Anelastic Creep-Induced Structural  
Anisotropy in a Metallic Glass  
J. Haimovich and T. Egami  
to be submitted to Scripta Met.

31. Microscopic Mechanism of Anelastic and Plastic Deformation:  
Computer Study  
Y. Suzuki and T. Egami  
in preparation.
32. Structural Anisotropy Produced by Anelastic Deformation of  
Amorphous Alloys: X-Ray Diffraction Observation  
Y. Suzuki, J. Haimovich and T. Egami  
in preparation.

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